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## Giant optical nonlinearity of heterostructures with InP self-assembled quantum dots

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**Abstract.** Quantum dots (QD) are an attractive object for study due to their expectable nonlinear properties. We studied nonlinear reflection of heterostructures with self-assembled InP QDs by use of specially designed pump-probe technique of high sensitivity. We discovered that built-in electric field present in our structures considerably enhances the effect of the QD excitation and allow us to study it at rather low level of the excitation power.

### Introduction

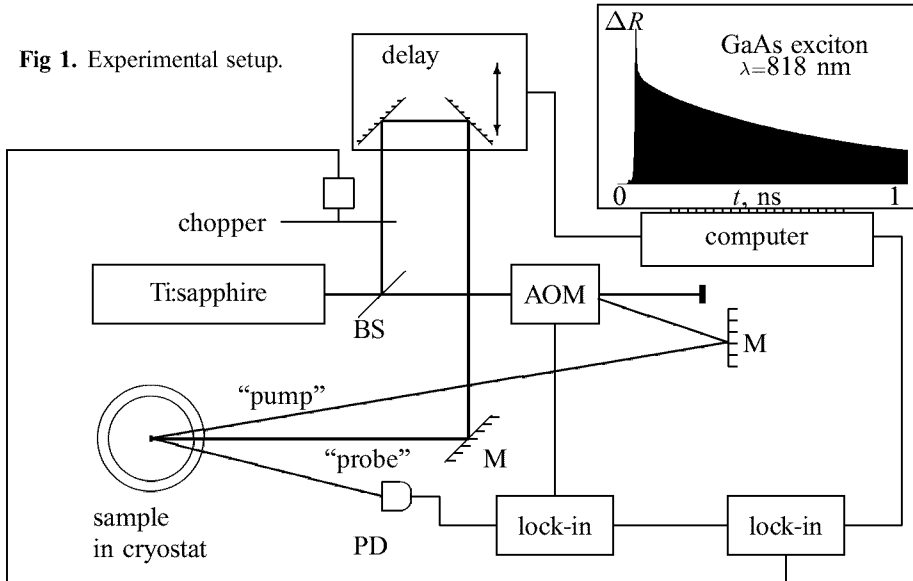
Among the other semiconductor nanostructures quantum dots (QDs) attract considerable interest because of their delta-like density of states. This peculiarity leads to the high nonlinearity both in optical and electrical properties of the dots. A number of the interesting manifestations of the optical nonlinearity is really found by few groups [1–3]. The investigations in this field are of particular interest because they promise wide range of applications in the fields of optics, opto-electronics and electronics.

### 1 Experimental

This work is devoted to study nonlinear reflection of the heterostructures with the InP self-assembled QDs. The structures were grown by gas source molecular beam epitaxy (GS MBE) on  $n^+$  GaAs substrates. They contain one layer of InP QDs between  $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$  barrier layers grown on a GaAs buffer layer. We studied a few structures with the different thicknesses of the barrier layers and the QD sizes.

Pump-probe experiments were performed by means of the setup which is schematically drawn in Fig. 1. This setup includes a femtosecond Ti:sapphire laser "Tsunami" (power source "Millenia", frequency 82 MHz, pulse duration 0.1–1 ps) which is tunable from about 700 to 850 nm. Amplitude modulation of the pump and probe beams at different frequencies, optical phase shift between them and lock-in detection of the signal modulated at the differential frequency let us to avoid noises from the scattered light and achieve high sensitivity of detection of nonlinear reflection whose detection limit is about  $10^{-7}$  times of the linear reflection.

Fig 1. Experimental setup.

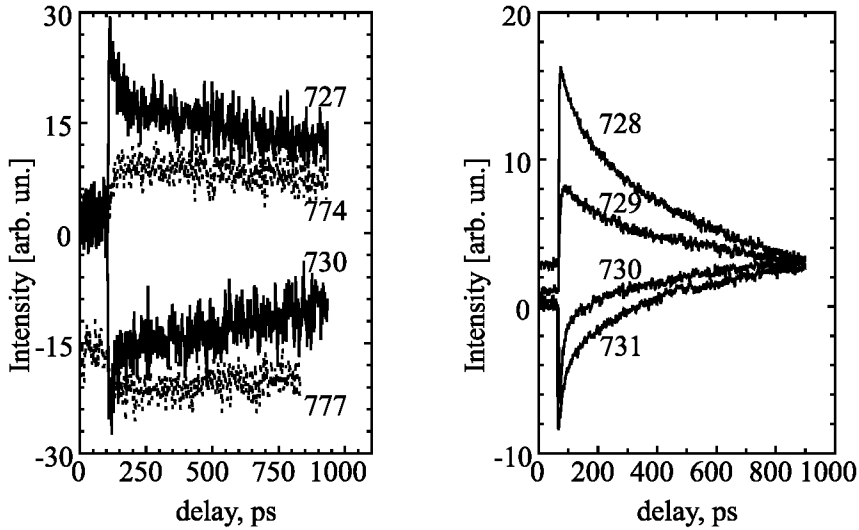


## 2 Results and discussion

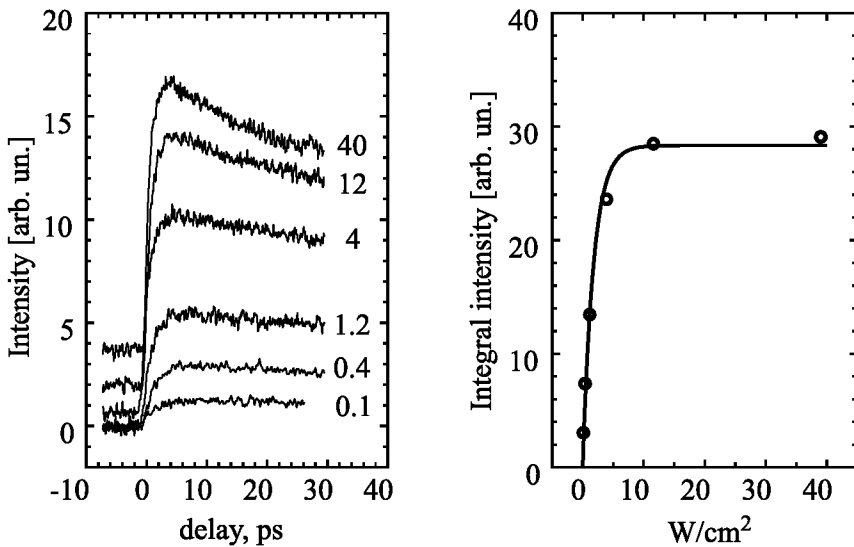
In Fig. 2 the typical time dependences of a pump-probe signal are presented. This signal exhibits short risetime (equal to the time resolution of our setup) and exponential decay with a few characteristic times.

In the spectral region of the InP QD photoluminescence (PL), 710–760 nm for a sample QDO1505 where the PL intensity is at the level of 0.1 times of the peak intensity, main decay time is about 0.8 ns. This decay is presumably due to the energy relaxation in the QDs. Also there is a long component of the signal with characteristic time much longer than 12 ns that is a time separation between successive light pulses in our setup. It is seen as a constant pedestal and caused probably by the space separation due to the diffusion of the photogenerated carriers in the GaAs layer produced by pump pulses. Under the stronger excitation a fast decay time component of about 30 ps is also observed. Most probably, it is due to energy relaxation of hot carriers in the GaAs buffer layer. In the wavelength region beyond the QD PL band, a contribution of the main decay component decreases rapidly and is hardly seen up to the GaAs bulk exciton spectral region.

Pump-probe signal as is seen in Fig. 2 has an interesting spectral dependence which looks like intense oscillations with almost constant period of 6.5 nm. We studied this phenomenon in details in [4] and found that this behaviour is due to built-in electric field which is present in our structures. It leads to Franz-Keldysh oscillations (FKO) in the reflection spectrum. A study of the spectral behaviour of FKO and their dependence on pump power together with an analysis of time evolution of the signal provides us strong evidence that we really detect a change of reflection due to optical excitation of QDs in spite of the low optical density of QDs. We found that the built-in electric field



**Fig 2.** Pump-probe signal for the sample QDO1505 at the different spectral points (marked by wavelength in a units of nm) under the pump power density of  $P = 0.1 \text{ W/cm}^2$  (left panel) and  $100 \text{ W/cm}^2$  (right panel).



**Fig 3.** Left panel: pump-probe signal under different pump power densities (in a unit  $\text{W/cm}^2$ ); right panel: power dependence of the integrated signal (circles) and its fitted line by (1) with  $I_0 = 28$  and  $P_0 = 1.8 \text{ W/cm}^2$ .

can drastically enhance the effect of the QD excitation and allows us to study it at very low excitation levels.

A dependence of the pump-probe signal on the pump power is most important for the study of the optical nonlinearity. It is shown in Fig.3 for the spectral point of 727 nm. One can clearly see that these dependences exhibit saturation which can be fitted by the simple equation

$$I = I_0 \left( 1 - e^{-\frac{P}{P_0}} \right). \quad (1)$$

Saturation power  $P_0$  is only about  $1.8 \text{ W/cm}^2$ . Inverse of the areal density of QDs estimated by means of atomic force microscopy is  $0.1 \times 0.1 \mu\text{m}^2$ . This means that  $1.8 \text{ W/cm}^2$  corresponds to only 9 photons per dot per pulse. The saturation power has the same order of magnitude for all studied heterostructures in the spectral points inside the region of QD absorption. We do not observe any quick saturation of signal in the spectral region beyond the QD PL band.

### 3 Conclusion

The heterostructures with InP self assembled QDs are studied by the pump-probe method. We found that the build-in electric field which is present in the studied structures allows us to detect a changes of reflection due to optical excitation of QDs under extremely low pump power. In the spectral region of QD absorption, a pump-probe signal exhibits a quick saturation. Saturation power is only about  $2 \text{ W/cm}^2$  or 10 photons per dot per pulse. This fact evidently shows the giant nonlinearity of the QDs.

#### Acknowledgements

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